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Under the auspices of the JANNAF Combustion Subcommittee, a workshop on the subject "Influence of Gas-Phase Chemical Kinetics on the Low-Pressure Ignition and Flamespreading in Solid Propellants" was held at Hampton, VA, on 23 and 24 October 1986 in conjunction with the 23rd JANNAF Combustion Meeting. The objectives of the workshop were to a) force a synergistic interaction between those whose measurements have provided evidence for the participation of chemical kinetics in low-pressure solid propellant burning and those who are striving to simulate these events with interior ballistic models and b) address problems of anomalous ignition behavior in gun propelling charges. The workshop participants a) reviewed the evidence for the participation of chemical kinetics in the low-pressure burning of solid propellants, b) reviewed model simulations which include finite-rate kinetics, and c) determined which model simulations and further experimental characterizations would be most fruitful. This report documents the workshop.

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I. INTRODUCTION

Under the auspices of the JANNAF Combustion Subcommittee, a workshop on the subject "Influence of Gas-Phase Chemical Kinetics on the Low-Pressure Ignition and Flamespreading in Solid Propellants" was held at Hampton, VA, on 23 and 24 October 1986 in conjunction with the 23rd JANNAF Combustion Meeting.

The stated objectives of the workshop were to a) force a synergistic interaction between those whose measurements have provided evidence for the participation of chemical kinetics in low-pressure solid propellant burning and those who are striving to simulate these events with interior ballistic models and b) address problems of anomalous ignition behavior in gun propelling charges.

The issue areas were to a) review the evidence for the participation of chemical kinetics in the low-pressure burning of solid propellants, b) review model simulations which include finite-rate kinetics, and c) determine which model simulations and further experimental characterizations would be most fruitful.

The justification for the workshop was stated as follows: Very long ignition delays (tens to hundreds of milliseconds) often accompany the low-pressure ignition event encountered with artillery charges; however, measured heat input to the propellant suggests that ignition should take place in just a few milliseconds. Further, high-speed movies of ignition and flamespreading in transparent howitzer simulators show first luminosity (following that of the igniter itself) often to be in the gas-phase regions of ullage downstream from the propellant bed; first propellant combustion with visible flame within the bed itself is often considerably downstream of the ignition source. Significant gas-phase chemistry is also suggested by the occasional, vigorous combustion event (e.g. breechblow) which accompanies extremely long ignition delays exhibited in firings of cold-conditioned charges. Recently, nitramine gun propellants, particularly low-vulnerability (LOVA) compositions, have revealed, on occasion, large disparities between calculated and experimental performance; significant flame-zone gas-phase reactions have been postulated as an explanation. Modeling efforts have begun to address these considerations. New interior ballistic codes include provision for several species and several reactions. Available input data from the measurements community are often unsuitable, however. Progress in this area depends critically on constructive interactions between the modeling and the measurements communities.

The following people were active participants in the workshop:

George Keller, U.S. Army Ballistic Research Laboratory, APG, MD
Carl Roller, U.S. Army Armament Research, Development, and Engineering Center, Dover, NJ
Ludwig Stiefel, U.S. Army Armament Research, Development, and Engineering Center, Dover, NJ
Martin Summerfield, Princeton Combustion Research Laboratories, Inc., Monmouth Junction, NJ
Martin Miller, U.S. Army Ballistic Research Laboratory, APG, MD
Tim Parr, Naval Weapons Center, China Lake, CA
Channon Price, Naval Weapons Center, China Lake, CA
Paul Gough, Paul Gough Associates, Portsmouth, NH
William Vienna, Naval Ordnance Station, Indian Head, MD
Arpad Juhasz, U.S. Army Ballistic Research Laboratory, APG, MD

The workshop presentations can be roughly divided into observations of systems or system simulators, observations in the laboratory, and modeling status.

II. SYSTEM OBSERVATIONS

Keller presented observations of T.C. Minor and L.M. Chang which were made in the transparent gun chamber simulator at the Ballistic Research Laboratory. Using transparent and translucent chamber-diameter tubes, the simulator makes it possible to use high speed photography and X-rays to observe primer functioning, igniter functioning, and low-pressure flamespreading phenomena in full sized charges for 105-mm and 120-mm tank guns, for a 5"/54-caliber Navy gun, and for 155-mm howitzers. For the case of charges that used M1 propellant, interesting observations were made of luminosity at the forward end of the charge that followed the primer functioning but definitely preceded the ignition of the main propellant. In some of these situations, enough pressure was generated to cause motion of the forward bag of a multiply-bagged howitzer charge. The observation that this luminosity could be severely reduced by filling the chamber with nitrogen gas, rather than having it full of ambient air, seemed to indicate that gas phase chemistry was responsible for the luminosity. Later observations with low vulnerability propellant (LOVA) revealed very long (tens of milliseconds) ignition delays, followed by what appeared to be normal functioning, which again suggested that gas-phase reactions could be responsible for ignition delays in systems using LOVA propellant. For this discussion, "ignition delay" is understood to be the time from the primer firing to the beginning of rapid chamber pressurization.

The discussion following the presentation emphasized the fact that ignition delays were certainly not unique to LOVA, but that if there were a weak ignition system, LOVA propellant would certainly exacerbate the problem. Suggestions were made to include firings in oxygen as well as the ones in nitrogen.

Roller reported on tests of new primers that have been specifically designed to ignite LOVA propellants more efficiently. LOVA was designed to resist thermal initiation; a complication of this design has been that it exhibits marginal ignition characteristics with standard Benite primers. As a result, some rounds with LOVA propellants have had long ignition delay times, poor ballistic uniformity, and poor propulsion efficiency. Laboratory studies suggested that oxygen as an igniter product would be beneficial. Thus, primers using "Oxite," which is designed to yield free oxygen at equilibrium, have been developed. The latest in the series uses a modified Oxite, which yields 7% oxygen at equilibrium. The ballistic tests with these primers have been good, and ballistic variability has been significantly reduced. Not surprisingly, the main propellant is also more completely burned up. Another feature of the new primers, of less interest to this workshop, was that they also pressurize differently, and that improves their performance with LOVA propellants. The studies concluded that an oxygen-producing igniter material is more effective for LOVA propellant than non-oxygen-producing material of similar energy and flame characteristics.

The ensuing discussion emphasized the need both for reactions in the condensed phase and for gas phase reactions in the foam layer (which would be a function of pressure) in a flame model of the combustion of solid propellant. It has been observed that first light and first measurable pressure are not coincident for nitramines.

Stiefel then presented the "highlights" of a meeting of the JANNAF Panel on ignitability properties of LOVA propellants, which was held at Naval Ordnance Station, Indian Head, MD

on 12 August 1986. The panel considered emerging LOVA compositions, primer output characterization methods, generalized propellant bed ignition studies, fundamental ignition studies, and ballistic modeling. The newest of the emerging LOVA's is HELOVA, in which energetic plasticizers with high temperature stability have been substituted for ATEC, and in which a titanate or zirconate coupling agent has been added. The processing agent gives improved processability and mechanical properties while also yielding reduced vulnerability. The advantages of HELOVA are higher impetus, higher solid density, higher loading density, lower temperature coefficient, lower barrel wear, and increased survivability. A very interesting device for studying primer output in detail was described. It provides pressure-time data of events within the primer, and it permits observing the flamespread in the primer. A large number of tests to explain the observed temperature coefficients in LOVA propellants were listed. Flamespreading mechanisms that might be important to the performance of LOVA propellant grains have been investigated in detail, and it has been found that the pressure at which flame penetrates the perforation is inversely proportional to the perforation diameter. Fundamental propellant ignitability studies on LOVA propellants were described, including flow ignition studies, laser ignition studies, and hot fragment conductive ignition. Considerable success in modeling the global ignition kinetics for hot fragment conductive ignition was reported.

Summerfield then presented an extensive review of the rocket literature, extending back many years, which supports the importance of the role of detailed chemical kinetics in the flame of a burning solid propellant. Practically, detailed chemical kinetics affects a) efficiency of combustion, b) stability of a rocket motor, and c) the dynamics of extinguishment. It has application to a) rocket motors utilizing metalized propellants, b) smokeless rocket propellants, and c) high energy rocket propellants.

For solid propellants, one can infer from the thickness of the dark zone a time constant of about 0.010 seconds. For a rocket, for which the core velocity is about 1000 ft/sec, heat release would take place about 10 feet downstream! Similar calculations for a gun propellant, for a pressure of, say, 10,000 psi, and a time of reaction of about 0.001 second (for a bimolecular reaction, the time of reaction goes as 1/p), then heat is released about 1 foot from the propellant surface. In this case, the gas phase flame zone, smoke, luminosity, and heat release are far downstream from sources. [See later comments by Kuo, however.]

In the Zeldovich theory, all the heat is released in a thin flame. For ammonium perchlorate (AP), theory matches observations. For double base propellants, that theory doesn't work. Slow gas phase reactions are going on, and, unless the volume is sufficient to let all gas phase reactions go to completion, to get steady state burning, all heat will not be released. There was no particular theory for this situation.

As reported in the Journal of Spacecraft, Vol 10, 1973, experiments were performed with transparent-walled rocket motors at 500 to 700 psi. The work was performed with four different propellants. One was nitrocellulose (NC) double base, but it did not contain NG, and it had an inert binder. There should have been a thin flame zone, but instead, a large dark zone was observed. The investigators added AP until the flame zone was all luminous. Recall now that the dark zone is the invisible portion of the visible flame in which there are appreciable slow reactions. Thin flames and AP go together. Summerfield suggested that one should, at first, assume a 50/50 ratio between heat release at the surface and heat release in the gas phase.

Previously, when one of Summerfield's students modeled flame chemistry, he included a limited number of species. The effort was based upon shock tube work, with other known

chemistry. With much faster computers now available, one could put in many more reactions. One would observe slow reactions still proceeding at the end of the calculation.

In an AGARD proceeding, Schoyer reported some work on solid propellants. He made an L-star rocket. Its behavior was easy to explain for AP, but completely confusing when double base propellant was used. The instability that then appeared was dominated at two different times by two processes, where the second process has to do with a second type of heat release in the gas phase. This work is still unexplained.

In work by Kubota, the length of the dark zone was observed to vary inversely with pressure. The nature of the flame for nitramine or double base propellant is fundamentally different from AP flame, for there is no "dark zone" with AP. Pictures of nitramine propellant combustion at 300 psi., 20 atmospheres, showed a thick flame with a large dark zone.

Work by Weaver, in which measurements are made of several molecules including CN using laser fluorescence to a spatial resolution of 20 μm, concludes that a highly unsteady process like interior ballistics is very hard to model and depends strongly on gas phase reactions.

In the discussion that followed this review, Kuo pointed out that the products of the combustion of solid propellant have to go through the boundary layer, where the flow velocity is less than that in the free stream. Thus the spacing may be overestimated in the rough estimates that were made earlier. Stiefel emphasized that it is the ignition event, not full-flame burning, in which slow gas phase reactions are important. Gough remarked that we now model double base propellants fairly well, but not LOVA. LOVA seems fundamentally different, and our lack of understanding of its behavior leads to problems in modeling it.

III. LABORATORY OBSERVATIONS

Martin Miller reported that, in the course of research on burning rate mechanisms of solid propellants, he and his colleagues have developed a semi-empirical method for characterizing the effective kinetics controlling the rate of heat release in the fizz zone (the "near-field" flame) of burning solid propellants. The method requires an experimental temperature profile through the zone of heat release. It assumes that the far-field flame zone does not influence burn rate, and further assumes a good measurement of temperature profile and the ability to pinpoint the location of the propellant surface. The same analysis could be adapted to characterize the kinetics of heat release in the visible flame zone provided the temperature could be profiled there. Techniques involving refractory thermocouples or laser Raman thermometry might be developed to provide the necessary calibration measurements. The applicability of these kinetics, derived from isobaric conditions, to gun chambers may be hindered by convective detachment of the visible flame from the burning grain and the attendant uncertainties associated with the mixing and dilution of the reactants. However, these issues might be put to the test by modeling and instrumenting the pressurization of a small closed bomb by propellant grains (or a single grain) of simple geometry.

Miller has observed that, unlike single base M10 and double base M9, triple base M30 has no dark zone, burning with a flame structure like AP. He notes that, at 50 atmospheres, Zenin observed staged combustion related to first luminosity at the downstream end of double base propellant.

In the discussion that followed, it was pointed out that turbulence may slow down -- or speed up -- reactions. Also, in the combustion of solid propellant in a gun charge, gases will have been percolating through the bed. Further, pyrolysis gases may have passed through no fizz zone, so that pyrolysis considerations may have to be included.

Several participants emphasized the fact that pyrolysis can be either exothermic or endothermic, and that it must be treated correctly for flame models to be successful.

Parr described his experiments using imaging Planar Laser Induced Fluorescence (PLIF) to measure species profiles during CO_2 laser ignition and steady state deflagration of HMX and aluminized nitramine high energy propellant (HEP's). All experiments were done at one atmosphere, in air, though the technique could be used at higher pressures. Time resolved PLIF images were obtained for NO_2, NO, CN, NH, and OH during ignition of both HMX and HEP's. CN and NH were observed to form at reasonably large heights off the sample surface in gas phase ignition kernels at finite delays, and then to reform into relatively thin flame sheets which "snap" back towards the sample surface. NO_2 and NO are initial products during laser ignition, beginning to form at minimum delay times and producing tall plumes until ignition occurs in the gas phase. After that, PLIF signals for both NO_2 and NO separate into an expanding shell and a solid, bell-shaped core close to the propellant surface. Thus it appears that NO_2 and NO are early decomposition products which are consumed in a flame separated significantly from the surface. The CN and NH are produced in this flame as transient radicals. OH is produced at the same delay as CN and NH, but it extends very far beyond the thin CN/NH flame front. Evidence was given that the flames are two phase, with particulates well above the surface, even for neat HMX.

The participants agreed that it would be exciting to see PLIF applied to LOVA combustion; some discussion followed concerning obtaining the propellant for the tests.

Juhasz then described the mobile combustion diagnostic fixture (MCDF), which is now under construction at the Ballistic Research Laboratory. MCDF is composed of a combustion chamber and an expansion chamber which are separated by a calibrated rupture disk. The entire device is mobile and is mounted on a trunnion so that it may be rotated at will. MCDF was designed with a sufficient safety factor that it can be operated in a laboratory with diagnostic facilities, even a laser laboratory, rather than having to be operated in a gun range. The combustion chamber and the expansion chamber have many diagnostic ports, which can be used with pressure gauges, windows for laser probes, or fiber-optic probes. The expansion chamber has additional ports for evacuation or backfilling before use and for sampling stable combustion intermediates. By using blowout disks with a range of pressures, one hopes to examine combustion intermediates as a function of pressure and shed light on the complicated kinetic processes involved in the combustion of solid (or liquid) propellants.

IV. MODELING STATUS

Price described a kinetics energetics modeling effort (for three propellants, including HMX) in which steady state burn rate data from three initial temperatures and arc image ignition data are fitted in order to deduce detailed reaction rates. He uses two reaction paths to final products,

A -> B -> C

and

A -> B' -> C,

having found that using only one reaction path just doesn't permit fitting the observed data. The A to B or B' reaction is the condensed phase reaction, with B or B' to C the flame zone reaction. If the condensed phase reaction is exothermic, it produces mostly formaldehyde. If endothermic, it produces mostly HCN. At higher pressures, he and his colleagues believe the condensed phase reaction is more and more endothermic. This method for arriving at global reaction rates caused much discussion.

Vienna summarized the TRESP code, which was written by Paul Gough. TRESP is a one-dimensional unsteady code which describes the thermal response of a reacting solid propellant. The code uses a one dimensional Fourier transport expression coupled with multiple Arrhenius (or absolute) chemical reactions, and the "hot end" boundary condition may take the form of variable heat flux or temperature, as input by the user. These options allow a wide variety of heating and reaction scenarios for investigation or verification of combustion kinetic schemes.

Vienna next presented a simplified combustion model for solid propellants in which the surface regression is controlled by a limiting surface temperature. An analogy of this qualification is an equilibrium phase change from liquid to gas where the liquid temperature is held constant by a vapor pressure law. It is significant that the solution of the equations describing heat transport (Fourier's + reaction heat) and reaction rate (absolute theory) for different temperatures reduces to an $r = a * P^n$ law. A vigorous discussion of the shortcomings of the model followed. The discussion ended with a consensus that if ignition and combustion of solid propellants is to be modeled at a reaction kinetics level, neither the gas nor the solid phase kinetics can be ignored.

Gough detailed the capabilities of the XNOVAK code, which includes detailed chemical reactions in propellant combustion. It is one of the latest is the family of NOVA codes, which are one-dimensional, two-phase-flow interior ballistic codes. In XNOVAK, the gas phase was replaced by a multiphase mixture of both gas- and condensed-phase combustion products, the latter consisting of either liquid droplets or solid particles. The mixture of combustion products is treated as homogeneous, in that all its constituents are assumed to have the same velocity and temperature. Chemical reactions may occur in the mixture of combustion products. There may be, as the code is presently written, as many as ten different species and ten different reactions. All reactions are assumed to have rates governed by a general Arrhenius law. Each reaction is assumed to proceed in the forward direction only, consistent with the assumption that the reactions are global and not fundamental reactions. Reactions are assumed to proceed with time constants of the same order as interior ballistic functioning, so that a stiff integration package is not included. The user provides a list of species and their properties, and the initial mass fractions of each of the species. He provides the igniter function as a mass flux, as before, but now also specifies the chemical composition of the igniter products, which is assumed to be the same at all positions and times. As the surface of the propellant heats to ignition temperature and begins to regress, intermediate propellant combustion products of a type specified are produced, and a fraction of the total combustion energy of the propellant is released. Later reactions transform these intermediate products to final combustion products and release the remainder of the available energy. Since all the species move with time, the final release of energy may or may not take place at or near the original propellant location.

Gough expressed a desire to add a natural transition from near field flame to far field flame and to add proper heat feedback to the model.

Keller reported calculations he has performed with XNOVAK over the last two years, which were presented at both the 1985 and the 1986 JANNAF Combustion meetings. The calculations show that it is plausible for gaseous products of the igniter to react in the gas phase with the intermediate products of combustion of the solid propellant to speed the conversion of intermediate products to final products. In that case, the energy associated with the conversion to final products would be released earlier in the ballistic trajectory, and that would lead to increased weapon system ballistic performance. While these calculations do not prove why oxygen-rich primers work better with LOVA propellants, they do help substantiate the most likely hypothesis.

V. CONCLUSIONS

The workshop participants formulated the following conclusions from the workshop:

1. Transparent simulator results suggest energy-releasing gas-phase reactions at sites away from the igniter stimulus.

2. Primer chemistry matters.

3. History shows slow reactions probably important in rockets and, in the ignition phase, for guns.

4. Slow reactions should be important for nitramines and double base propellant, but only nitramines have obvious problems. The flame structures look the same. We may have overlooked clues.

5. There is laboratory evidence for staged heat release in the burning of nitrate ester (except M30) and nitramine solid propellant.

6. A reliable process for deducing global kinetics from temperature profiles has been worked out and validated.

7. Simple flamespreading tests might prove to be useful to sort propellants with respect to relative behavior.

8. Diagnostic techniques have come a long way in the last few years.

9. Progress is being made in the area of kinetics.

10. Transient combustion / ignition is a strong function of kinetics.

11. There are now interior ballistic codes which include rudimentary chemistry models.

12. Interior ballistic code provides plausibility argument for Oxite primer / LOVA performance observations.

VI. SUGGESTED FUTURE ACTIVITY

The workshop participants formulated the following list of activities that should take place in the future:

1. More one-dimensional transparent simulator observations are needed for LOVA, JA2, etc.

2. More analyses of primer tests are needed.

3. The effects of reactions on closed bomb measurements should be determined.

4. MCDF could be used to find that pressure at which the expected equilibrium products predominate.

5. The thermocouple - temperature measurement technique needs to be established.

6. Laser-based diagnostics should be applied to the flames of a wide variety of propellants -- there are many candidates.

7. The use of a variety of propellants illuminates proposed models. Workers should not use just one; using only one can deceive!

8. Interior ballistic codes need near-field flame model.

9. Near-surface and sub-surface reactions' effects on interior ballistics require elucidation.

11. Thermal response studies using several models are needed.

VII. REQUIREMENTS FOR THE STUDY OF FUNDAMENTAL CHEMISTRY

The workshop participants formulated the following list of reasons that detailed chemical reaction rates and mechanisms must be determined:

1. They help sort out the applicability of models.

2. Ultimately, they could help predict the performance of a new propellant.

3. Ultimately, detailed chemical paths could be reduced to global kinetics.

4. They may help diagnose the source of problems in deviant propellant lots.

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The following references to some of the work which was presented at the workshop are included here for the benefit of the reader.

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